



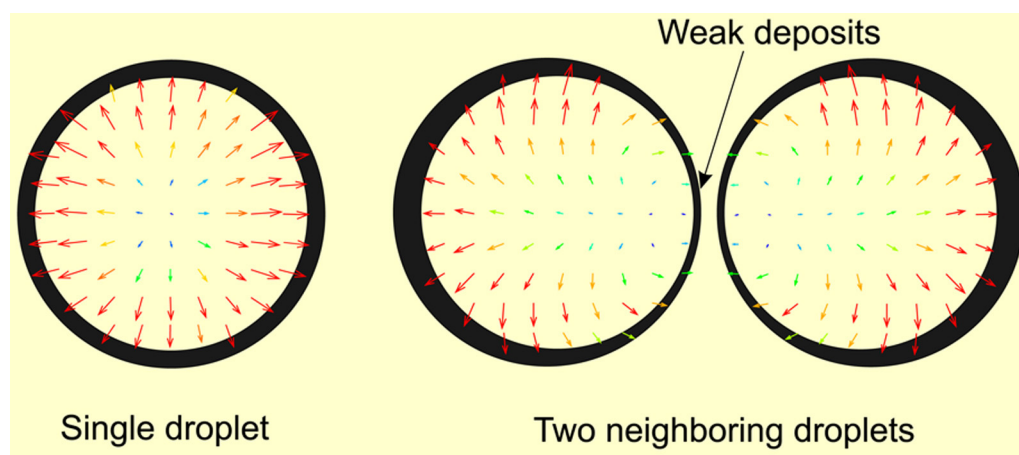
## Deposition pattern of interacting droplets



Tapan Kumar Pradhan, Pradipta Kumar Panigrahi\*

Department of Mechanical Engineering, Indian Institute of Technology Kanpur, Kanpur 208016, India

### GRAPHICAL ABSTRACT



### HIGHLIGHTS

- Drying pattern of droplets is influenced by droplet interaction.
- Non-uniform deposition pattern is observed for two droplets drying adjacent to each other.
- Presence of a neighboring droplet influences the evaporation flux on the droplet surface.
- The convection pattern is asymmetric in two droplets configuration.

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### ABSTRACT

A colloid droplet having pinned contact line forms a ring pattern after drying. The ring pattern of a single droplet shows uniform deposit along the contact line. When two droplets dry adjacent to each other, the deposition pattern is influenced by the presence of the neighboring droplet. The deposition pattern at the nearest region of the two droplets shows weak deposit compared to the rest of the region. The drying pattern in two-droplets configuration is related to the convection phenomena inside the drying droplet and the influence of the neighboring droplet. We have experimentally studied the effect of a neighboring droplet on the convection pattern inside the evaporating droplet using  $\mu$ -PIV technique to explain the influence of transport phenomena on the drying pattern. COMSOL simulation has also been carried out for understanding the evaporation process. The evaporation rate from the nearest region of the two droplets is lower compared to the rest of the region. Therefore, less number of particles are transported to this region due to low fluid flow causing less deposits in this region. The flow field inside a droplet in the

\* Corresponding author. Tel.: +91 5122597686; fax: +91 5122597408.

E-mail addresses: [tapankp@iitk.ac.in](mailto:tapankp@iitk.ac.in) (T.K. Pradhan), [panig@iitk.ac.in](mailto:panig@iitk.ac.in) (P.K. Panigrahi).

presence of a neighboring droplet shows asymmetric convection pattern unlike single droplet where flow field is symmetric. This convection behavior correlates well with the evaporation flux distribution on the droplet surface and drying pattern.

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## 1. Introduction

Drying of a droplet containing suspended particles forms deposition pattern on the substrate after drying [1]. The deposition pattern is found in drops of aqueous salt solution [2,3], DNA solution [4,5], blood [6], suspended nanoparticles [7,8] etc. Evaporation driven pattern deposition has many practical implications like two-dimensional crystallization [9], surface coating [10–12], DNA chip manufacturing [5], biosensing of protein [13], disease diagnosis [6,14] etc. The famous coffee ring [15] pattern occurs due to the movement of particles towards the contact line. Evaporative flux at the contact line of a droplet is more as compared to the top of the droplet [15,16]. Fluid is convected towards the contact line to replenish the evaporative loss at the contact line [15]. The particles get convected along with the fluid and deposited at the edge forming coffee ring pattern [15]. Pattern deposition depends on many factors like particle size [17,18], droplet size [18], nature of substrate surface [19,20], particle shapes [21,22], flow pattern inside droplet [15,23,24], presence of surfactant and additives [25,26], substrate temperature [22], droplet composition [27] etc.

Flow pattern inside a droplet significantly affects the deposition pattern of the droplet. Outward flow inside droplet leads to the movement of particles towards the contact line forming ring like deposits [15]. In the presence of Marangoni convection, where fluid flows towards the center of the droplet, reverse effect of coffee ring happens and the particles get deposited at the center [23]. Marangoni flow can also be used to form ordered hexagonal and stripe-like nanoparticle patterns [28]. Evaporation condition from the droplet surface affects the flow pattern leading to different deposition pattern [29,24]. For edge enhanced evaporation, fluid flows towards the center forming ring like deposits and for center enhanced evaporation, fluid flows towards the top of the droplet leading to uniform deposits [24]. Internal flow generated inside the droplet by electrowetting can suppress the coffee ring forming uniform deposits [30].

All the above studies have been carried out in single droplet configuration. However, the hydrodynamics inside an evaporating droplet is significantly affected by the presence of a neighboring droplet. Carles and Cazabat [31] observed that a PDMS droplet can be propelled by the presence of a volatile trans-decalin droplet. Cira et al. [32] study the mobility of two neighboring droplets of water-PG mixture. They observed both attraction and repulsion between the two droplets depending upon the PG concentration. When two drops placed nearer to each other dry simultaneously, they form weak deposits at the region of greatest proximity due to lower evaporation at this region [29]. Chen and Evnas [33] reported the formation of arched structure of dried droplet when the droplet dries in the presence of another droplet nearer to it. In this paper, we have studied the effect of neighboring droplet on the hydrodynamics inside a droplet of pinned contact line and relate it to the deposition pattern. We have experimentally studied the particle convection during drying process of a droplet with and without the presence of a neighboring droplet and the effect of neighboring droplet on the deposition pattern.

## 2. Experimental methods

DI water with conductivity of 1.05  $\mu\text{S}/\text{cm}$  has been used as the working fluid and the volume of each droplet has been set equal

to 0.3  $\mu\text{L}$ . Fluorescent polystyrene particles of 2  $\mu\text{m}$  diameter with concentration of (0.018%) volume fraction have been used as seeding particles. Water containing the seeding particles is sonicated for 15 min in an ultrasonication bath (Crest Ultrasonics model-230D) for proper mixing of the seeding particles and to break the agglomerates of particles. Droplets of water containing these seeding particles are placed on a clean microscope glass slide. The glass slide is made of soda lime glass. The glass slide is thoroughly cleaned by sonicating in acetone. The roughness of the glass surface is measured by Qualitest surface roughness tester (Model TR100). Surface roughness ( $R_a$ ) of the glass surface is equal to 0.1  $\mu\text{m}$ . The droplet with required volume is dispensed on the glass surface using a micro pipette. The droplet forms contact line diameter of approximately 1.8 mm. The contact angle of the droplet is equal to  $38^\circ \pm 4^\circ$ . DropSnake plugin in Image J [34] is used to measure the contact angle. After drying, the polystyrene particles form a deposition pattern. These fluorescent polystyrene particles also act as tracer particle for velocity measurement using particle imaging velocimetry (PIV). We have studied the flow pattern during the drying process and deposition pattern after drying of a single droplet and two droplets.

Flow inside the droplet during drying process is studied using micro PIV technique. Images for the PIV measurements have been captured in confocal microscope arrangement (Fig. 1). The fluorescent particles are illuminated by a laser source of wavelength equal to 488 nm. The emission from the particles is captured by the PMT present in the confocal microscope. The background noise is minimal as a confocal microscope uses pin hole. The separation time between two consecutive images are kept at 1.315 s. The size of each image is equal to  $512 \times 512$  pixels. The images are processed by using DynamicStudio V1.45 software to get the velocity vector field information inside the droplet.

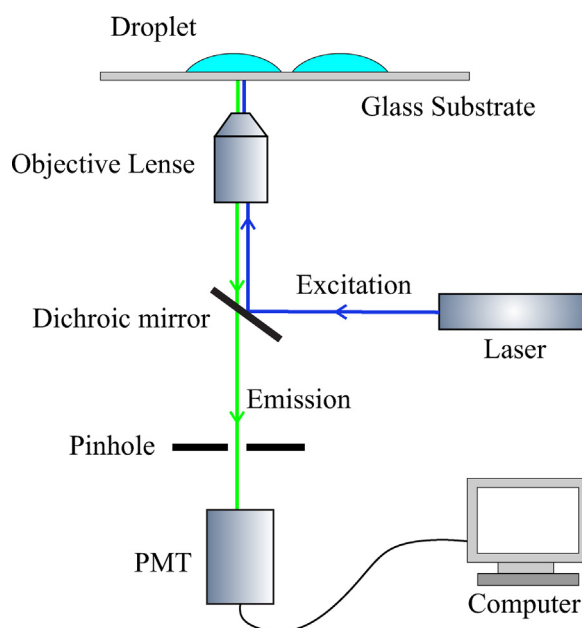


Fig. 1. Experimental arrangement for studying the convection pattern inside evaporating droplets.

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